Chapter 16
Diffusion and Transportation Dynamics of ^{137}Cs Deposited on the Forested Area in Fukushima After the Fukushima Daiichi Nuclear Power Plant Accident in March 2011

Nobuhito Ohte, Masashi Murakami, Takahiro Suzuki, Kohei Iseda, Keitaro Tanoi, and Nobuyoshi Ishii

Abstract A massive amount of radioactive substances, including cesium-137 (^{137}Cs), emitted from the disabled nuclear power plant has deposited on the forested areas in the northeastern region of Honshu Island, Japan after the Fukushima Daiichi nuclear power plant accident. Forests in these regions are particularly important not only for the forest products industry but also for source areas of drinking water and for residential environments. To clarify the mechanisms of diffusion and export of ^{137}Cs deposited on the forested ecosystem, we initiated intensive field observations in a small catchment, including forest and farmlands, in the Kami-Oguni River catchment in the northern part of Fukushima Prefecture. The following expected major pathways of ^{137}Cs diffusion and export were investigated: (1) transportation by water movement of dissolved and particulate or colloidal forms through hydrological processes and (2) diffusion through the food web in terrestrial and aquatic ecosystems of forests. Preliminary findings indicated the following: (1) most of the ^{137}Cs was discharged as suspended matter, and particulate organic matter appeared to be the most important carrier of ^{137}Cs. High water flow generated by storm accelerated the transportation of ^{137}Cs from the forested catchments. Estimation of ^{137}Cs export thus requires precise evaluation of the high flow acceleration during storm events. (2) Because litter and its detritus may form the biggest pool of ^{137}Cs in the forested ecosystem, ^{137}Cs diffusion occurs more rapidly through the detritus food chain than the grazing food chain. Most predators have already ingested ^{137}Cs, particularly in aquatic environments. An urgent question is when and how ^{137}Cs diffuses through grazing food chains and how rapidly

N. Ohte (✉) • K. Iseda • K. Tanoi
The Graduate School of Agricultural and Life Sciences,
University of Tokyo, Tokyo 113-8657, Japan
e-mail: nobu@fr.a.u-tokyo.ac.jp

M. Murakami • T. Suzuki
Graduate School of Science, Chiba University, Chiba 263-8522, Japan

N. Ishii
National Institute of Radiological Sciences, Chiba 263-8555, Japan

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this process occurs. To elucidate or predict these phenomena, the mechanisms of ^{137}Cs release from litter and soil organic matter need to be clarified.

Keywords ^{137}Cs • Food web • Forest • Fukushima Daiichi nuclear power plant • Hydrological processes

Abbreviations

^{137}Cs	Cesium-137
^{131}I	Iodine
MAFF	Ministry of Agriculture, Forestry and Fisheries
MEXT	Ministry of Education, Culture, Sports, Science and Technology

16.1 Introduction

The Fukushima Daiichi nuclear power plant accident in March 2011 emitted 1.5×10^{17} Bq of iodine-131 (^{131}I) and 1.2×10^{16} Bq of cesium-137 (^{137}Cs) into the surrounding environment [Ministry of Education, Culture, Sports, Science and Technology (hereafter, MEXT) and Ministry of Agriculture, Forestry and Fisheries (hereafter, MAFF) 2012]. These radioactive substances, including ^{137}Cs, were deposited on the forested areas in the northeastern region of Honshu Island, Japan. Forests in these regions are particularly important for humans not only for their association with the forestry industry but also for the source areas of drinking water and for residential environments. The first phase of government surveys and investigations showed that the major portion of deposited ^{137}Cs was trapped in the canopy and litter layer at the soil surface (Hashimoto et al. 2012; MEXT and MAFF 2012). ^{137}Cs has been shown to be easily adsorbed onto clay minerals and soil organic matter (Kruyts and Delvaux 2002), which can be transported by eroded soils and particulate and dissolved organic matter through hydrological channels, streams, and rivers (Fukuyama et al. 2005; Kato et al. 2010; Wakiyama et al. 2010). Dissolved ^{137}Cs, which is relatively free from soil adsorption, can also be taken up by microbes, algae, and plants in soil and aquatic systems. This form of ^{137}Cs will eventually be introduced into soil insects and worms, fishes, and birds through the food web (Fig. 16.1).

To precisely describe the mechanisms of diffusion and export of ^{137}Cs deposited on the forested catchment, field observations of two primary but different pathways were conducted. One was the transportation of ^{137}Cs through hydrological flow paths as particulate or dissolved substances. The other was its diffusion through the food web of plants and animals in forested and aquatic ecosystems.

In this chapter, we present the concept, method, and implementation of the field experiments and also report the results of preliminary investigations 15 months after the accident. The main focus of the field observations was the flow of materials not only of ^{137}Cs itself but also of related compounds through the continuum of forested

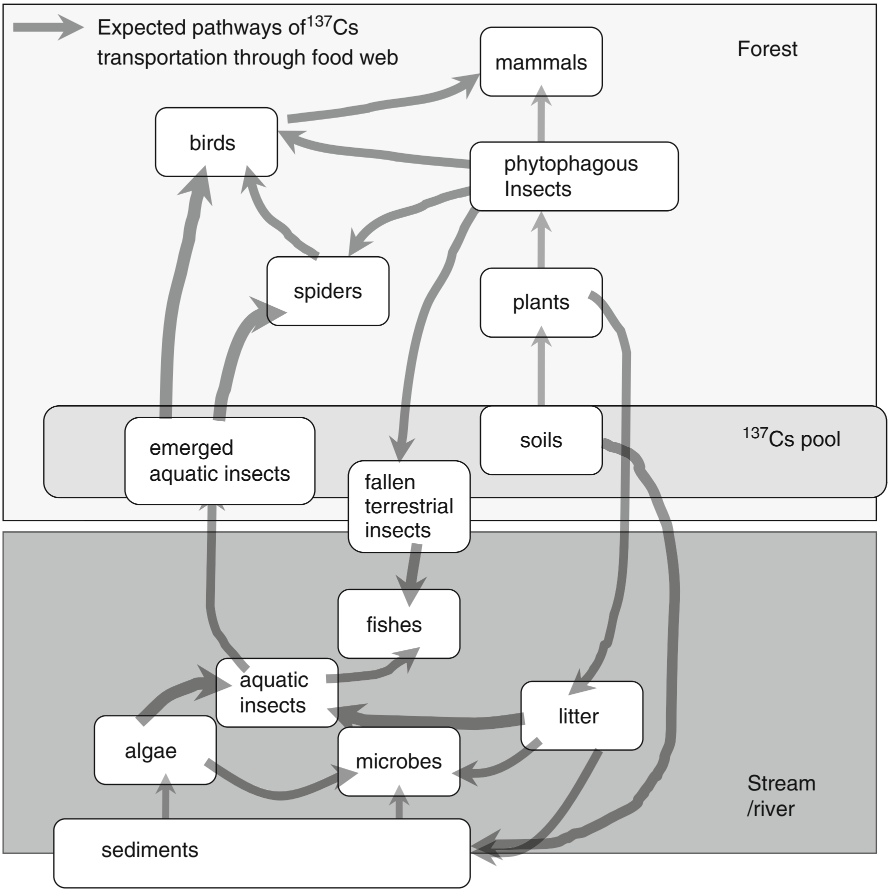

Fig. 16.1 Conceptual diagram of the pathways of cesium-137 (^{137}Cs) transfer thorough the food webs in aquatic and terrestrial ecosystems (based on Ohte et al. 2012)

and stream ecosystems. We are currently investigating how ^{137}Cs is transported by water and suspended materials and how it diffuses through the food webs in the forest–stream ecological continuum.

16.2 Materials and Methods

16.2.1 Study Site

The study was conducted at the Kami-Oguni River catchment in the northern part of Fukushima Prefecture. The area is located approximately 50 km from the Fukushima Daiichi nuclear power plant (Fig. 16.2a). According to a radioactivity survey report by MEXT that was performed using aircraft survey devices, the air

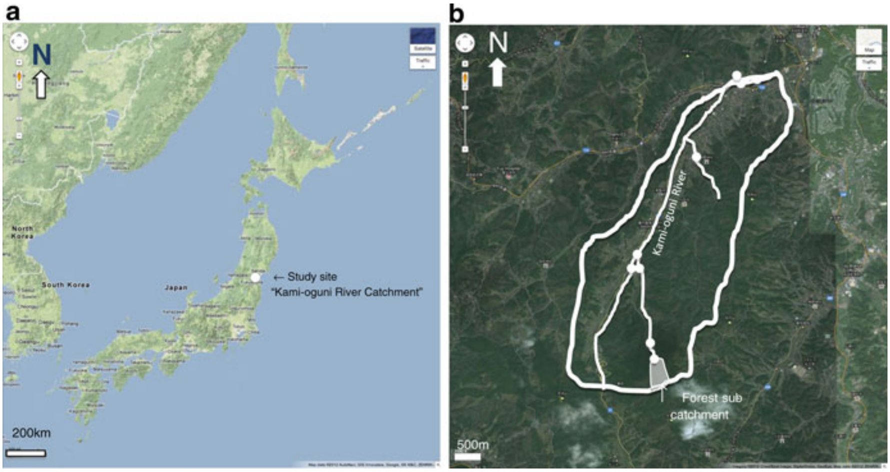

Fig. 16.2 (**a**) Location of the study site. (**b**) Observation facilities in the study catchment

dose rate in this region was 1.9–3.8 μSv/h and the total deposition rate of ^{137}Cs was 300–600 kBq/m^2 at June 28, 2012 (MEXT 2012).

The study catchment predominantly comprises forests and paddy fields, and the dominant tree species are broadleaf deciduous trees because the forested regions were mainly used as coppices until the 1960s (Watanabe, personal communication). Some parts were used as Japanese cedar and cypress plantations for timber production. The agricultural land also includes orchards producing peaches and Japanese persimmons.

In 2011, brown rice produced in several paddies in this area was found to have high radiocesium concentrations exceeding the provisional regulation level set by the Ministry of Health, Labour and Welfare of Japan. Based on this survey, MAFF decided to prohibit rice plantation during the 2012 season (MAFF 2011). The Date City government and local farmers only planted test plantations to investigate the rate of ^{137}Cs uptake by rice and the efficacy of countermeasures (applying potassium and materials that adsorb ^{137}Cs) in several selected paddies.

The upstream part of the study catchment is mostly forested, whereas the farmland areas, including the paddy fields, are mostly found in the middle to downstream parts of the catchment.

16.2.2 Survey

16.2.2.1 Forested Catchment

Within the forested parts of the study catchment, a small subcatchment was selected for intensive observation and sampling (Fig. 16.2b). Three rectangular plots

(20 m × 20 m), including two deciduous stands and one cedar plantation, were studied to determine the processes involved in ^{137}Cs pools and flows.

To describe the distribution and sizes of the ^{137}Cs pools found within the subcatchment, we collected periodical samples of litter, plants, soil, and stream water and measured the ^{137}Cs concentration of each sample.

To determine fluxes in ^{137}Cs deposition, movement within the subcatchment, and discharge from the catchment, we measured the water flux and ^{137}Cs concentration for every factor associated with ^{137}Cs movement, including rain, throughfall, stem flow, and streamwater discharge. The water discharge rate from the subcatchment was continuously measured using a partial flume with a water level gauge. Throughfall and stem flow water were collected from both the secondary deciduous stands and the cedar plantation.

Samples were collected from all terrestrial food web members, including litter and living leaves, soil worms, insects, lizards, and snakes.

16.2.2.2 River Transect

Water samples were also collected from nine points along the Kami-Oguni River to elucidate the riverine distribution of ^{137}Cs (Fig. 16.2b). The total length of the river transect was 7 km from the outlet of the forested subcatchment. Confluence locations were generally selected as sampling points. Samples were collected from members of the aquatic food web, including litter detritus, benthic algae, aquatic macrophytes, benthonic organisms, and fish.

16.2.3 Analysis

Water samples were filtered with a glass fiber filter (Whatman GF/F, $\phi = 0.7$ μm) immediately after sampling. The ^{137}Cs concentration of suspended matter present on the filter was measured. Condensing treatment using a germanium semiconductor detector is required to measure the dissolved ^{137}Cs concentration. This will be performed in the next phase of the study. Samples of litter and organisms were dried in an oven, ground into a powder, and homogenized. Germanium semiconductor detectors at the University of Tokyo and the National Institute of Radiological Sciences were used for measuring ^{137}Cs concentrations of all the samples.

Carbon and nitrogen concentrations and the isotope composition ($\delta^{13}C$ and $\delta^{15}N$) of the samples were also determined. Isotope data were used to evaluate the trophic levels of sample organisms according to the methodology reported by Lajtha and Michener (1994).

16.3 Preliminary Results and Discussion

16.3.1 ^{137}Cs Export by Hydrological Processes

The observed spatial distribution of ^{137}Cs in suspended matter along the Kami-Oguni River in June 2012 is shown in Fig. 16.3c. A large storm event, a typhoon, occurred in the study area from June 19 to 20 (Fig. 16.3a, b). The increased runoff caused by this storm event exported much of the suspended matter from the source areas of the stream, such as the riparian zones of the forested areas (riparian topography is rarely found in agricultural areas because of their well-developed artificial drainage systems). The ^{137}Cs concentration in the suspended matter increased significantly during the storm event; however, it was diluted by the water discharged from the farmlands in the middle to downstream parts of the catchment. The contribution of the forested catchments located in the uppermost regions was significant.

The ^{137}Cs concentration decreased rapidly over the next day (June 20). This quick response suggests that the mechanism of ^{137}Cs discharge was highly dependent on rapid flows such as the overland flow that occurred in the riparian zones of the forested areas. While this response was rapid, it should be noted that the ^{137}Cs concentration in the suspended matter was higher than that before the storm event even after 1 week.

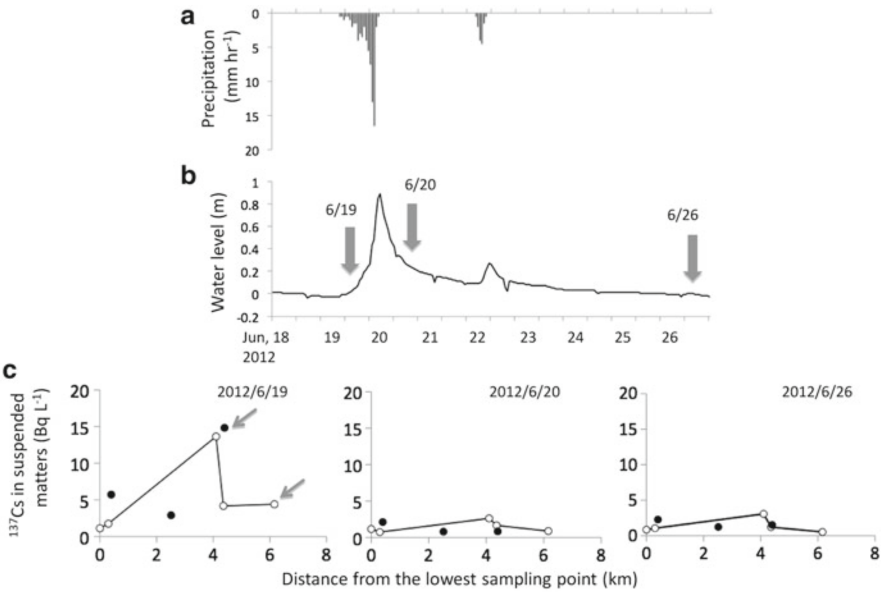

Fig. 16.3 Observed spatial distribution of cesium-137 (^{137}Cs) concentration in suspended matter along the Kami-Oguni River in June 2012. (**a**) Precipitation at Iidate, (**b**) water level of the Hirose River at Ohzeki (downstream of the Kami-Oguni River), (**c**) ^{137}Cs concentration of suspended matter. The *arrows* on the panel of 2012/6/19 indicate the influxes from the forested catchments (based on Ohte et al. 2012)

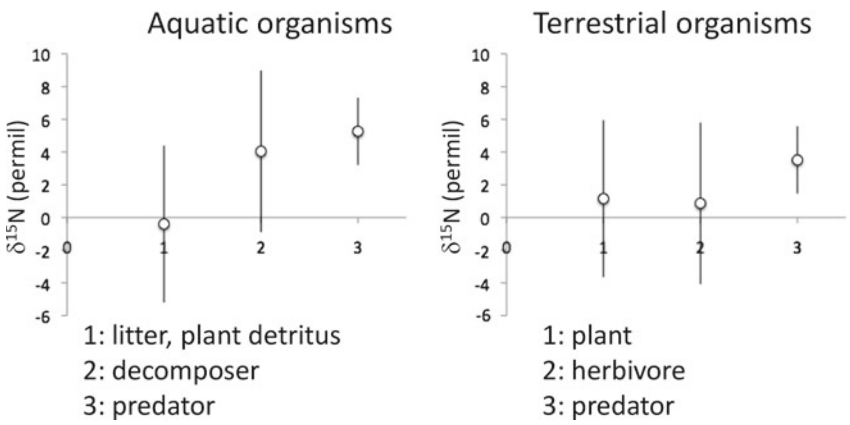

Fig. 16.4 $\delta^{15}N$ value for each expected trophic level of functional groups of aquatic and terrestrial organisms (based on Ohte et al. 2012)

It is generally accepted that the amount of suspended matter increases during conditions of high flow in headwater streams during storm events. It is also often observed that sources of suspended matter are the stream bank and the riparian zone surface. Our data suggest that suspended matter plays the most important role in the export of ^{137}Cs from the forested catchment. This interpretation is also supported by a study on other forests in Fukushima, which found that most of the ^{137}Cs pool remained in litter and decomposing organic materials found on the forest floor and soil surface (Hashimoto et al. 2012).

These findings indicate that high flows generated by storm events accelerate the transportation of ^{137}Cs by water from forested headwater catchments (Fig. 16.3). Precise estimation of the extent of ^{137}Cs export from forested catchments requires a realistic evaluation of high flow acceleration during storm events.

16.3.2 ^{137}Cs Diffusion Through the Food Web

The $\delta^{15}N$ values increased with the expected trophic levels of functional groups of aquatic and terrestrial organisms (Fig. 16.4). The ^{137}Cs concentrations in these trophic groups are shown in Fig. 16.5. The ^{137}Cs concentration in litter was significantly higher than that in the other groups. The concentration in mushrooms was also high, showing a similar tendency to that seen in a previous report (MEXT and MAFF 2012).

The ^{137}Cs concentration in aquatic organisms was generally higher than that in terrestrial herbivores, which clearly indicates that the majority of the aquatic primary consumers feed on litter detritus, whereas the primary consumers of the terrestrial food web mainly feed on living plant tissues. In other words, the fastest diffusion of ^{137}Cs occurs through the detritus food chain, and it is dominant in the food web of aquatic organisms. In contrast, in terrestrial organisms, herbivorous insects did not ingest ^{137}Cs, although the ^{137}Cs concentration of predators was

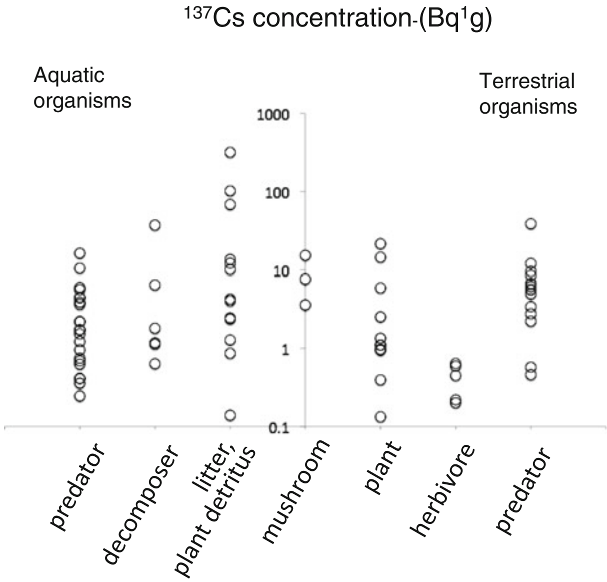

Fig. 16.5 Cesium-137 (^{137}Cs) concentrations of three trophic groups of aquatic and terrestrial organisms (based on Ohte et al. 2012)

significantly high. This suggests that many plants have not yet absorbed ^{137}Cs and that terrestrial predators such as lizards and snakes may consume some decomposers found in the shallow soil layers, e.g., angleworms and galleyworms. Figure 16.5 also indicates that ^{137}Cs is substantially diffused to primary consumers but has not completely reached yet high-level predators, particularly in the aquatic ecosystem.

An urgent question is when and how ^{137}Cs diffuses through the grazing food chain and how rapidly this process occurs. To predict this phenomenon, mechanisms of ^{137}Cs release from litter and soil organic matter as well as the ^{137}Cs absorbing behavior of plants need to be clarified.

16.4 Future Studies

Since the Chernobyl nuclear power plant accident, the dynamics of radioactive substances has been monitored in many forested ecosystems in Russia, the Baltic countries, and Scandinavia. Some of these have continued long-term ecosystem monitoring (Fesenko et al. 1995; Realo et al. 1995; Lehto et al. 2008). In southern Finland, for example, it was reported that the Chernobyl fallout remained on site, with more than 3,000 Bq/kg being found in the uppermost (0–3 cm) humus layer of forest soil (Ylipieti et al. 2008).

At present, in northeastern Japan, including Fukushima, the largest pool of fallout ^{137}Cs has been found in the litter layer of deciduous forest floors as well as in

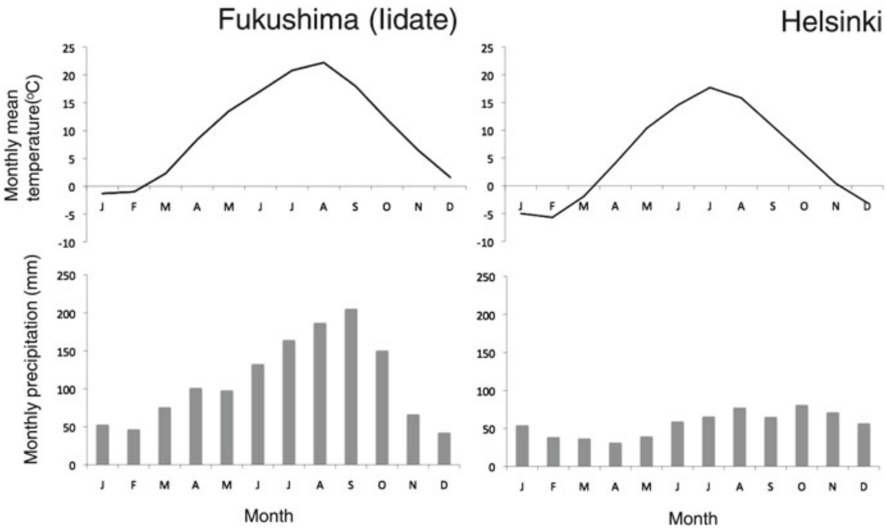

Fig. 16.6 Monthly mean air temperature and precipitation in Helsinki and Fukushima (Iidate town)

the canopies of evergreen coniferous forests. Therefore, the urgent and most important objective of future studies is to predict how fast ^{137}Cs moves from relatively fresh litter to the humus or mineral soils found beneath the litter layer and also how it is adsorbed to these soils. Although the Scandinavian case studies suggested high retention of fallout in humus, there are geographical differences between Scandianvia and Japan, including differences in climate and geology.

Figure 16.6 shows the mean monthly air temperature and precipitation for Helsinki and Fukushima (Iidate town). The most important point to note is that the total precipitation in Fukushima is twice that in Helsinki and that ~60% of the total precipitation falls in June, July, August, and September. This is a typical seasonal variation caused by the Asian monsoon system. High-temperature summers with high precipitation also accelerate not only the decomposition of litter on the forest floor but also the transportation of detritus and dissolved organic matter by hydrological pathways. Thus, mobilized ^{137}Cs may be transported more easily into the deeper soils and exported by streams in Fukushima than in Scandinavia.

In addition, the geological and geomorphological characteristics of Fukushima forests are substantially different from those of Finland. Japanese geological settings are generally affected by active orogenic movement. Uplift and erosion activities are much more active than those in Scandinavia and the northwestern part of the Eurasian continent. This may accelerate the erosion and export of surface materials from the forest floor because of the high precipitation rate in summer (Ohte and Tokuchi 1999).

Thus, we have to be careful when referencing information obtained from Russia, Ukraine, and Scandinavia because of environmental differences. For comprehensive comparison between Japan and these countries, intensive and long-term monitoring of various forested ecosystems in the northeastern parts of Japan is required.

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